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**MODELLING, SIMULATION AND CONTROL OF ETHYL ACETATE REACTIVE DISTILLATION COLUMN USING CHEMCAD**

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**ABSTRACT**

The modelling, simulation and control of a reactive distillation column for the production of ethyl acetate from acetic acid and ethanol has been studied using commercially available process simulation software, CHEMCAD<sup>®</sup>. The effects of key operating variables such as reflux ratio, distillate rate, feed tray location and activity model on the composition of ethyl acetate in the distillate at steady state were studied. Steady state simulation results show that if a double feeding strategy is adopted, acetic acid must be fed to the tray 2 while ethanol must be fed to tray 12 in order to obtain optimum composition of ethyl acetate in the liquid phase. Also, a reflux ratio of 12 resulted in optimum composition of ethyl acetate in the liquid phase. Increasing the distillate rate resulted in an increase in the composition of ethyl acetate in the liquid phase. Using the steady state results as an initial state, dynamic simulations were run for the purpose controlling the reboiler pressure and ethyl acetate purity. For pressure control, the Ziegler-Nichols (ZN) tuning method was used to tune the controller parameters resulting in an optimal value of integral square error (*ISE*) of 587.85 and corresponding values of 0.13 and 7.05 for proportional gain and integral time constant ( $K_c$  and  $\tau_i$ ) respectively. A simulation exercise such as presented in this work is very important and is increasingly becoming popular. This is because simulation of mathematical models of chemical processes is safe, cheap and less time demanding. This has also been helped by the development of computers with high computational capabilities.

**Keywords;** Steady-state, Dynamics, Control, Reactive distillation, Ethyl acetate

**INTRODUCTION**

Reactive distillation is a process integration technique that incorporates separation and reaction in a single unit. The reaction normally occurs in the liquid phase or occasionally at the surface of a solid catalyst in contact with the liquid phase<sup>[1]</sup>. This process takes place purely at the equipment level and no new functional relationship between both operations is introduced<sup>[2]</sup>. This means that the reaction does not affect the separation operation neither does the separation operation affect the reaction. Popular applications of reactive distillation include technologies producing ether such as MTBE, ETBE, and TAME. Others include hydration of ethylene oxide to mono-ethylene glycol, and some instances of hydrogenation of dienes and aromatic compounds<sup>[3]</sup>. There are a host of other processes in which reactive distillation may become a potentially useful option. These include esterifications such as ethyl acetate from ethanol and acetic acid<sup>[4-7]</sup>, alkylation of aromatics and aliphatics such as ethylbenzene from ethylene and benzene<sup>[8]</sup>, cumene from propylene and benzene<sup>[9,10]</sup>,

decomposition of ethers to high purity olefins and so on. Reactive distillation systems have the capacity to reduce capital and operating costs especially for cases where the reactions are reversible and/or when azeotropes are formed making separation by conventional distillation systems consisting of separate reactor and distillation units difficult and expensive<sup>[11,12]</sup>. Other benefits of using reactive distillation include increased selectivity, conversion, speed and efficiency of operation, potential to overcome chemical equilibrium limitations, potential to reduce energy consumption through energy integration, generation of less waste and fewer by products. In spite of above stated benefits, reactive distillation cannot be used for every process that requires reaction and separation in a single unit. It is constrained by the fact that it is not an attractive option for operations carried out under supercritical conditions. It is also not very suitable for gas-phase reactions and for reactions that require high temperatures and pressures<sup>[1]</sup>. Notwithstanding these perceived constraints, there are many documented successes recorded in terms of implementation and commercial capacity with regards to the industrial application of reactive distillation<sup>[2]</sup>. Taylor & Krishna<sup>[13]</sup> also reported that the applications of reactive distillation in the chemical and petroleum industries have increased rapidly over the past decade.

A lot of the initial research effort in the area of reactive distillation was channelled mainly in the direction of development of design procedures under steady state conditions. In the last decade, a lot of attention has been focused on the dynamic modelling of reactive distillation though ETBE and MTBE production has enjoyed more attention<sup>[11]</sup>. Taylor and Krishna<sup>[13]</sup> presented an outstanding review on the modelling of reactive distillation. Recently, the control of reactive distillation columns has received some attention. Most of the works done in this area are on the production of ETBE. Vora & Daoutidis<sup>[14]</sup> studied the dynamics and control of a reactive distillation set up for ethyl acetate production. The initial set up they proposed was such that both reactants were fed to the column via different trays thus allowing for a higher conversion and purity to be obtained at the steady state. This performed better than the conventional set up which involved feeding both reactants to the column via the same tray. An analysis of the column dynamics was performed to reveal the existence of fast and slow dynamics corresponding to liquid flow rates, composition and holdups. The dynamic analysis of the column was followed by comprehensive analysis of possible control strategies. The control configuration adopted was that which could achieve constant energy requirements and bottom flowrates. These were studied in terms of the effect of the reflux rate (a manipulated variable) on the product purity (a controlled variable). Based on dynamic simulations, model-based linear and nonlinear feedback controllers along with conventional proportional integral (PI) controllers were designed and their performances evaluated. By running dynamic simulations, Sneesby *et al*,<sup>[15]</sup> offered general recommendations for the control of ETBE producing reactive distillation columns and also recognised the importance of attending to the issue of control at the early stages of the design process. Bock, *et al*.<sup>[16]</sup> proposed a control strategy for a reactive distillation column that incorporates a recovery column. Kumar & Daoutidis<sup>[17]</sup> studied the dynamics and non-linear control of an ethylene glycol reactive distillation column. They developed a control

system that performed well with stability in the high purity region. Sneesby *et al.* [18] considered the advantage of a control configuration for an ETBE producing column with composition and conversion as double control objectives. Luyben [19] compared the dynamic behaviour and steady-state economics of two alternative reactive distillation systems. Monroy-Loperena *et al.* [20] studied the non-linear control of an ethylene glycol reactive distillation column and proposed a robust PI control strategy. Georgiadis *et al.* [11] presented an experimentally validated hybrid dynamic. The model was used to study the interactions of design and control and operability in an ETBE producing reactive distillation column.

In this work, the production of ethyl acetate from the esterification of acetic acid with ethanol in a reactive distillation column is studied. A number of challenges are encountered when adopting reactive distillation. Bock *et al.* [21], Chang & Seader [22] explained that these challenges are related to the difficulty in achieving high ethyl acetate purity. Notwithstanding these challenges, this system has the merit of the availability of numerous experimental data in the literature for the purpose of results validation. Simulations were run using CHEMCAD® to determine the effect of process parameters such as reflux ratio, distillate rate and location of feed stage on the steady state composition of ethyl acetate in the distillate. Control of the reboiler pressure and ethyl acetate purity in the distillate was studied by running dynamic simulations.

## PROBLEM DEFINITION

The system under consideration consist of two feed streams of consisting of a saturated liquid acetic acid and ethanol. For the conventional configuration, both streams are fed to the column as a saturated liquid mixture via the sixth stage while for the case of double feed both streams are fed to the columns via two different stages. The column is made up of 13 trays and reaction takes place in all 13 trays of the column, with sulphuric acid as catalyst. The mixture is highly non-ideal mainly due to the presence of alcohol, acid and water. Five normal azeotropes exist in this mixture which makes the separation of pure components very difficult. The Non Random Two Liquid (NRTL) activity model was adopted for calculating the vapour-liquid-equilibrium (VLE) of this mixture. It incorporates a vapour phase model that can be used up to moderate pressures, and have the Poynting correction included in the liquid fugacity coefficient calculation. It can also handle any combination of polar and non-polar compounds and systems that exhibit a high degree of non-ideality.

## REACTIVE DISTILLATION COLUMN MODELLING

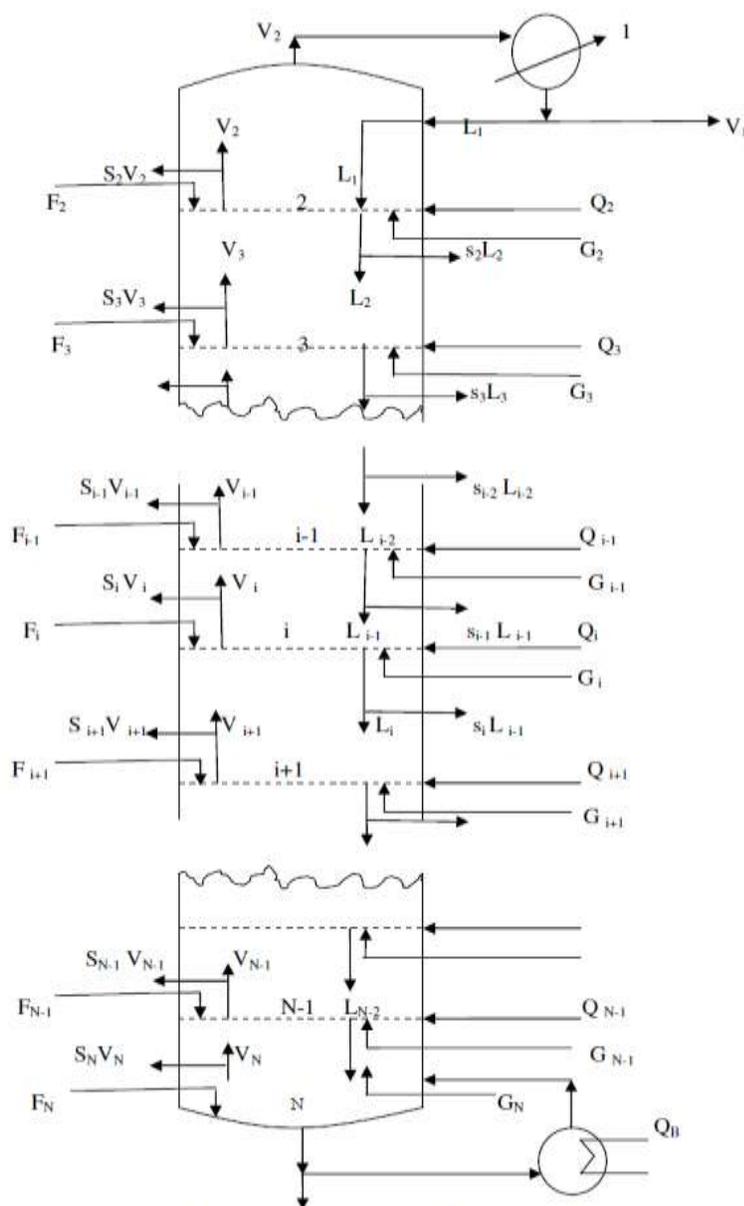
For the current study, a generic continuous N-stage reactive distillation column arranged in a countercurrent cascade was considered. A schematic representation of such a column separating  $C$  components is shown in Fig. 1. The stage numbering is from top to bottom such that stage 1 corresponds to a total condenser while stage  $N$  corresponds to a reboiler.

In developing the reactive distillation column model, the following assumptions were made:

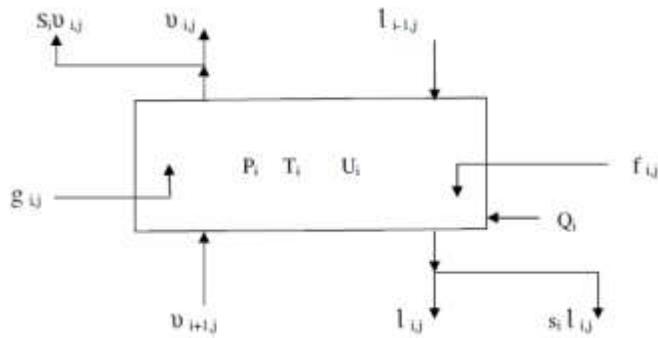
- The reaction occurs in the liquid phase.

- Each phase is perfectly mixed implying that liquid composition at each stage is homogeneous and equal to the composition of liquid leaving that stage.
- The vapour and liquid leaving any stage are in thermodynamic and mechanical equilibrium.
- Entrainment of liquid drops in vapour and occlusion of vapour bubbles in liquid are negligible

A generalised representation of a given stage is shown in Fig. 2.



**Figure 1:** Generic reactive distillation column configuration



**Figure 2:** General reactive distillation column stage

The expressions that describe the physical and chemical processes occurring on any given stage *i* are given in the following sections:

**Component Material Balances:**

$$M_{i,j} = (1 + S_i)v_{i,j} + (1 + S_i)l_{i,j} - v_{i+1,j} - l_{i+1,j} - f_{i,j} - g_{i,j} - U_i \sum_{n=1}^{NR} v_{j,n} r_{i,n} \quad (1)$$

Number of stages:  $i = 1, \dots, N$

Number of components:  $j = 1, \dots, C$

Number of reactions:  $n = 1, \dots, NR$

The rate of reaction is modelled by reversible power-law kinetic rate expressions.

$$r_{i, NR} = \sum_{p=1}^2 k_p \prod_{j=1}^C C_{i,j}^m = \sum_{p=1}^2 A_p \exp\left(\frac{E_p}{RT_i}\right) \prod_{j=1}^C C_{i,j}^m \quad (2)$$

$$C_{i,j} = \frac{l_{i,j}}{L_i v_i^L} \quad (3)$$

The terms are defined as follows:

$S_i$  Ratio of vapour draws off to primary vapour on stage *i*

$U_i$  Liquid holdup on stage *i*

$C_{i,j}$  Concentration of component *j* on stage *i*

$k_p$  Reaction rate constant for the *p*th term, (*p*=1 indicates the forward reaction and *p*=2 indicates the reverse reaction)

*m* Exponent of concentration

$A_p$  Pre exponential or frequency factor for the *p*th term

$E_p$  Activation energy of the reaction for the *p*th term

Because the material balance equations (1) are in the form out -minus- in, the term represents the moles of component *j* produced in the liquid phase. Therefore,  $v_{j,n}$  the stoichiometric coefficient for component *j* in the *n*th chemical reaction is positive if

component  $j$  is created by the  $n$ th reaction and negative if component  $j$  is consumed by the  $n$ th chemical reaction.

**Energy balances:**

$$E_i = (1 + S_i)H_i + (1 + S_i)h_i - H_{i+1} - h_{i-1} - h_{Fi} - H_{Gi} - Q_i \quad (4)$$

The terms are defined as follows:

- $E_i$  Residual energy for stage  $i$
- $H_i$  Enthalpy of the primary vapour leaving stage  $i$
- $h_i$  Enthalpy of the primary liquid leaving stage  $i$
- $H_{Gi}$  Enthalpy of total vapour phase on stage  $i$
- $H_{Fi}$  Liquid enthalpy of feed stream on stage  $i$
- $Q_i$  Heat duty at stage  $i$

The heat of reaction is not accounted for in the energy balance of equation (4) because enthalpies in the energy balance equation refer to the elements rather than to the components.

**Equilibrium Relationships:**

$$Q_i = K_{i,j} \frac{L_{i,j}}{L_i} v_{i,j} - v_{i,j} \quad (5)$$

$$K_{i,j} = \frac{y_{i,j}}{x_{i,j}} \quad (6)$$

$x$  and  $y$  represent the component mol fractions.

Equation (4) does not describe the total condenser which is also an equilibrium stage. If the reflux ratio is given, the top stage energy balance equation is expressed in terms of the reflux ratio as follows:

$$E_1 = L_1 - RV_1 \quad (7)$$

Specifications for equations (1-4) include the total number of stages, stage locations of all feeds, side streams, and heat exchangers, all stage pressures and liquid-phase holdup volumes; and complete specification of each fresh feed. This leads to a system of equations of  $N$  degrees of freedom as there are  $N(2C + 1)$  equations to be solved corresponding to  $2N(C + 1)$  variables. It is necessary to specify variables that are not dependent, hence if heat transfer rates for the  $N$  Stages are specified ( $N$  specifications), then the systems reduces to a square one of  $N(2C+1)$  equations and  $N(2C+1)$  variables. The equations are solved for  $2CN$  component molar flows and  $N$  stage temperatures. However, because condenser and reboiler duties are strongly dependent and can generally be specified independently over only narrow unknown ranges, other specifications are adopted for both stages. The solution of model equations is usually a complex process especially when the number of model equations and variables is large. Much effort has been channelled into rigorous solution strategies of model equations to obtain convergence. Over the last four decades, a lot of

numerical procedures for reactive distillation column models solution capable of implementation on digital computers have been developed and reported.

Many attempts are taken to simulate model equations rigorously using different methods by putting different criteria for convergence. Since 1970, calculation procedures suitable for implementation on digital computers, for rigorously solving the mass, equilibrium, and energy balances in multicomponent, multistage reactive-distillation columns have been reported. Chang & Seader <sup>[22]</sup> summarized the procedures for solving reactive distillation column models made up of combination of large sets of linear and nonlinear. These procedures can be divided into three groups viz: bubble point method which utilises tear variables, dynamic approach which utilises relaxation techniques and Newton-Raphson method which utilises an iterative approach. When control schemes are incorporated into the reactive distillation units, it introduces additional dynamics which, when coupled with dynamics of the unit, give rise to more complexity. It is usually very cumbersome when model solution is carried out manually. With the advent of fast digital computers with huge computation capacity and the development of commercially available process simulators, solving these model equations has become relatively easier to accomplish. One of such process used in this work is CHEMCAD<sup>®</sup>. It can easily cope with reactive distillation, a system which is highly non ideal because of the presence of a reversible reaction and the fact that Acetic Acid, Ethanol, Water, and Ethyl Acetate form a thermodynamically complex system with two-liquid phases and vapor phase association.

### **STEADY STATE SIMULATION**

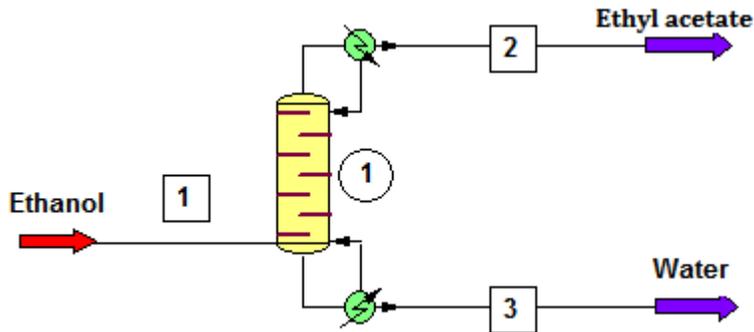
Steady state simulation involves the computation of the model variables in time  $t \rightarrow \infty$ , whereby the value of the variable becomes stable and time invariant. From a numerical point of view, steady state means that all derivatives with respect to time are equal to zero, i.e.  $d(\cdot)/dt = 0$ . Steady state simulation observes the behaviour of the system under steady state conditions. The reactive distillation column system under consideration in this work was simulated in CHEMCAD. Acetic Acid, Ethanol, Water, and Ethyl Acetate were selected as components while the Non Random Two Liquid (NRTL) property package was selected as the thermodynamic model for calculating the VLE of the non-ideal mixture. These specifications were made under the 'Thermophysical' tab in the CHEMCAD graphical user interface. The esterification reaction between ethanol and acetic acid was modelled as a kinetic reaction in CHEMCAD and added to the selected property package. The kinetic rate for the production of ethyl acetate used was adopted from Holland <sup>[23]</sup>. Different values of rate constants were used in order to account for catalysis.

#### ***Optimum location of Ethanol feed stage:***

For the single feeding strategy, the feed stream consists of a mixture of both acetic acid and ethanol and it is fed to the distillation column at the sixth stage (tray 6). Figure 3 shows the PFD for single feed strategy. The following specifications were made:

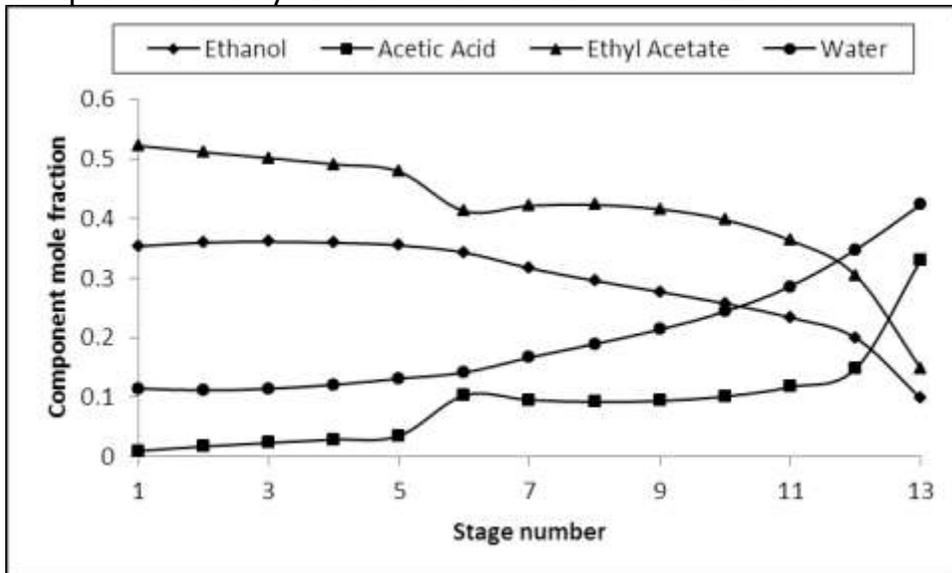
- Feed temperature: 70 °C

- Pressure: 1.2 bar
- Feed composition (molar flow):
- Acetic acid (CH<sub>3</sub>COOH): 24403.27 mol/h
- Ethanol (ETOH): 24403.27 mol/h
- Reflux ratio: 10
- Top stage efficiency: 0.75
- Bottom stage efficiency: 0.75



**Figure 3:** Single feed flow diagram in CHEMCAD user interface for RD  
 The result of the steady state simulation for the case of a single feed is shown below while the liquid composition profile is shown in Fig. 4:

Distillate rate: 19639.71 mol/h  
 Bottoms rate : 29166.82 mol/h  
 Composition of ethyl acetate in distillate: 0.5224  
 Composition of ethyl acetate in bottoms: 0.1486



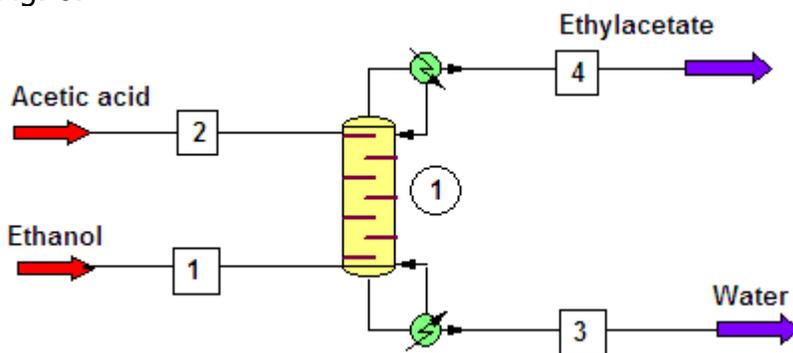
**Figure 4:** Liquid phase composition profile for single feed strategy

It is observed from Fig. 4 that the maximum composition of ethyl acetate is obtained at stage 1. The trend observed is such that the composition of ethyl acetate reduces gradually from stage 1 to stage 11 after which a sharp decline occurs between stages 11 and 13. Although the specifications adopted for the study are same as those of Kumar<sup>[24]</sup>, the results of this study as discussed are different. Comparisons with Kumar<sup>24</sup> are not included here because they used considerably different VLE as compared to those used here.

For the double feeding strategy, the following specifications were made:

- Feed temperature: 70 °C
- Pressure: 1.2 bar
- Feed composition (molar flow):
- Acetic acid stream : Acetic acid 24403.27 mol/h
- Ethanol stream : Ethanol: 43227.36 mol/h; Water: 7166.76 mol/h
- Reflux ratio: 10
- Top stage efficiency: 0.75
- Bottom stage efficiency: 0.75

In order to determine the optimum location of the ethanol feed stage, acetic acid was fed at a particular stage and kept constant while the feed stage of ethanol was varied. After doing this, the feeds were reversed and ethanol feed stage was kept constant while acetic acid feed stage was varied. The combination of ethanol and acetic acid feed stages that resulted in the maximum purity of ethyl acetate composition was adjudged to be the optimum feed stage and this combination was used for dynamics and control studies. Figure 5 shows the PFD for double feed strategy. Results of successive simulations as presented in Table 1 show that the best purity was obtained when acetic acid is fed at stage 2 and ethanol is fed at stage 12. The liquid composition profile corresponding to this feed combination is given in Fig. 6.



**Figure 5:** Double feed flow diagram in CHEMCAD user interface for RD

Table 1: Double feeding strategy for reactive distillation column

Acetic acid Feed tray	Ethanol Feed tray	Ethyl acetate mole fraction
Tray 2	Tray 4	0.5143
Tray 2	Tray 5	0.5335
Tray 2	Tray 6	0.5475
Tray 2	Tray 7	0.5578
Tray 2	Tray 8	0.5654
Tray 2	Tray 9	0.5711
Tray 2	Tray 10	0.5752
Tray 2	Tray 11	0.5784
<b>Tray 2</b>	<b>Tray 12</b>	<b>0.5807 (Optimum location)</b>
Tray 3	Tray 12	0.5675
Tray 4	Tray 12	0.5561
Tray 5	Tray 12	0.5471
Tray 6	Tray 12	0.5404

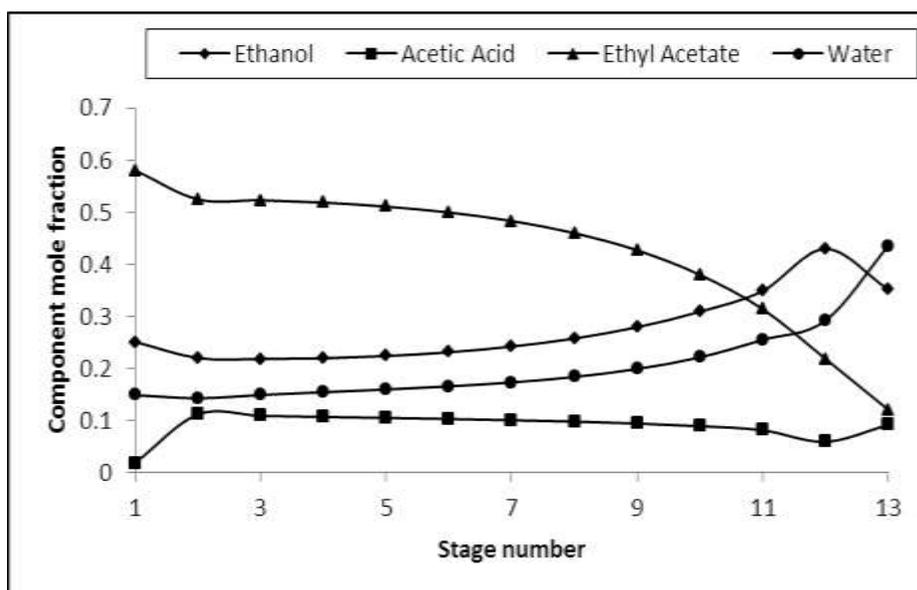
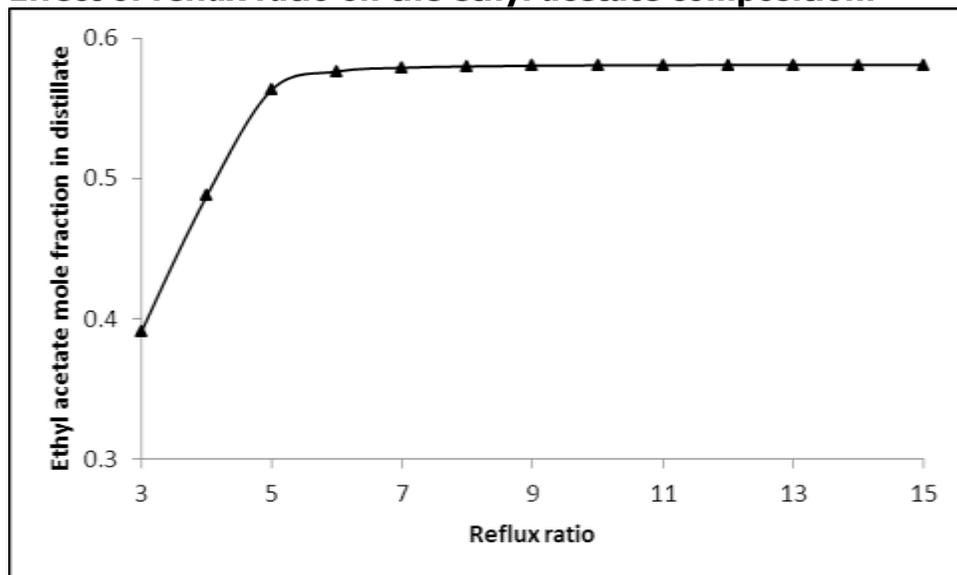


Figure 6: Liquid phase composition profile for double feed strategy

It is observed from Fig. 6 that the maximum composition of ethyl acetate of 0.5807 is obtained at stage 1 corresponding to the total condenser. The trend observed is such that the composition of ethyl acetate reduces in a gradual manner from stage 1 to stage 13.

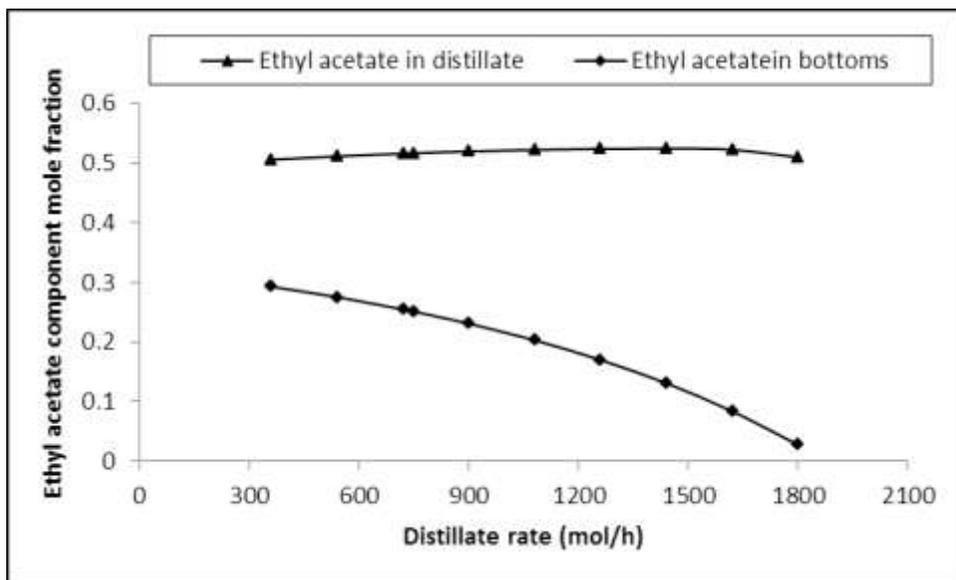
**Effect of reflux ratio on the ethyl acetate composition:**

**Figure 7:** Effect of reflux ratio on the ethyl acetate liquid phase composition

In order to determine how the reflux ratio affects the composition of ethyl acetate, steady state simulations of the ethyl acetate reactive distillation column were run at varying values of the reflux ratio within the range 3 to 15. Figure 7 shows the effect of reflux ratio on the liquid phase composition of ethyl acetate. The trend observed is such that the liquid phase composition of ethyl acetate increases from 0.3357 at a reflux ratio of 3 to a maximum of 0.5809 at a reflux ratio of 12. Increasing the reflux ratio beyond 12 had no effect on the liquid phase composition of ethyl acetate. This value of reflux ratio was adopted for subsequent simulations.

**EFFECT OF DISTILLATE RATE ON THE ETHYL ACETATE COMPOSITION**

By using the value of reflux ratio obtained from the previous section, and keeping other specifications constant, successive steady state simulations were run to determine the effect of distillate rate on the liquid phase composition of ethyl acetate. Figure 8 shows how the liquid phase composition of ethyl acetate in the distillate and bottoms vary with distillate rate.



**Figure 8:** Effect of distillate on the ethyl acetate liquid phase composition

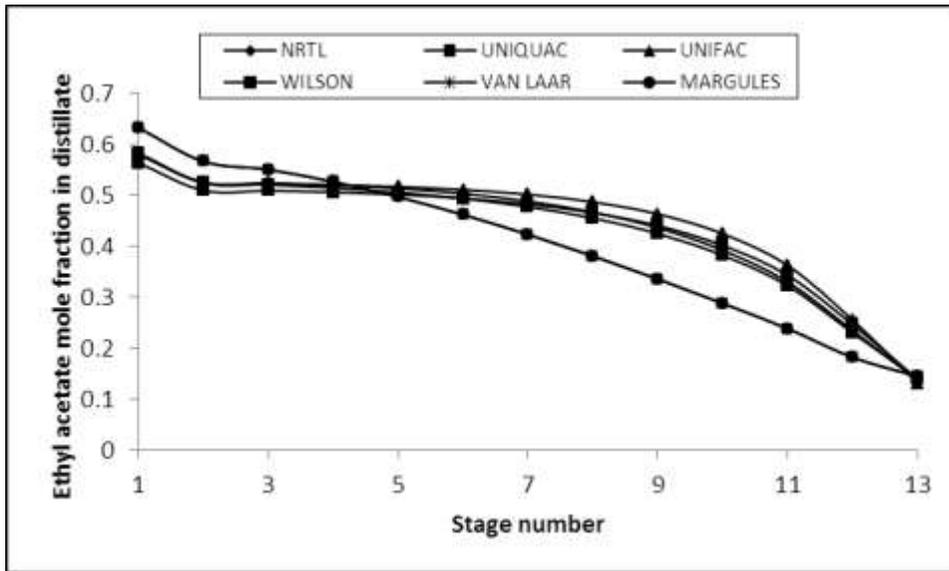
It can be observed from Fig. 8 that the liquid phase composition of ethyl acetate in the distillate increases slightly as the distillate rate was increased. Also the liquid phase composition of ethyl acetate in the bottoms decreased rather sharply as the distillate rate was increased. This observation is understandable in the sense that as the distillate rate is increased, the bottoms rate will decrease as a consequence of material balance. This means that more of the ethyl acetate will be present in the distillate as against the bottoms.

**EFFECT OF ACTIVITY MODEL ON THE ETHYL ACETATE COMPOSITION:**

In all the investigations carried out in this work before this section, the Non Random Two Liquid (NRTL) activity model was adopted for calculating the VLE of reactive mixture. In order to determine how the chosen activity model affect the values of ethyl acetate concentration in distillate and bottom, various activity models were tested and the result is shown in Table 2 and Fig. 9. Table 2 shows the composition of ethyl acetate concentration in distillate and bottom versus different activity models while Fig. 9 shows the liquid phase composition of ethyl acetate in the distillate.

Table 2: Effect of activity model on the composition of ethyl acetate

Activity Model	Ethyl acetate in distillate	Ethyl acetate in bottoms
NRTL	0.5809	0.1381
UNIQUAC	0.5643	0.1395
UNIFAC	0.5826	0.1313
WILSON	0.5835	0.1399
Van Laar	0.6334	0.1450
MARGULES	0.6334	0.1450



**Figure 9:** Effect of using different activity models on the ethyl acetate liquid phase composition

It can be observed from Fig. 9 that both the Margules and Van Laar activity models resulted in the same values for ethyl acetate liquid phase composition. The values calculated by the other models (NRTL, UNIQUAC, UNIFAC and WILSON) were not appreciably different from each other. This is probably because these four activity models are very accurate in representing vapour-liquid equilibria of non-ideal liquid mixtures. The WILSON activity model can describe the vapour-liquid equilibria of non-ideal liquid mixtures but the disadvantage in its usage is that it is not suitable for handling two phase systems.

### DYNAMIC SIMULATION

Dynamic simulation is usually the next step after simulating a process at steady-state. It is usually done for the purpose of control studies or to observe the behaviour or response of the process to a step change of the input variable. For the purpose of control studies, it is necessary to run dynamic simulations in order to determine the best control strategy to adopt. Dynamic simulation enables a better understanding of process performance and is a powerful tool for process plant optimisation, both at the operational and at the design stage. Also, steady-state operation is then better appreciated as the end result of a dynamic process for which all rates of change have eventually become zero. Based on dynamic simulations of the reactive distillation column system under consideration, the following control strategies were studied.

### PRESSURE CONTROL

Dynamic simulation for control purposes were carried out in CHEMCAD by using the steady state results obtained in previous sections as initial state. A proportional-integral (PI) controller was introduced to control the reboiler pressure. This was achieved by

manipulating the flowrate of steam to the reboiler. For a proportional-integral controller, we have the following expressions.

$$V_p(t) = V_p^{ss} + K_c \left( \varepsilon(t) + \frac{1}{\tau_i} \int_0^t \varepsilon(t') dt' \right) \quad (8)$$

Where  $V_p^{ss}$  is the valve position at steady state,  $K_c$  the proportional gain,  $\tau_i$  the integral time and  $\varepsilon$  the error? The controller error and reboiler pressure are related via:

$$\varepsilon(t) \equiv P_{set} - P(t) \quad (9)$$

Where  $P_{set}$  is the pressure set-point?

The integral square error is given as:

$$ISE = \int_0^t \varepsilon^2(t') dt'$$

(10)

Since valve position is bounded between 0 and 1, a Max (Min) equation was introduced to constrain the value of  $V_p$  between 0 and 1:

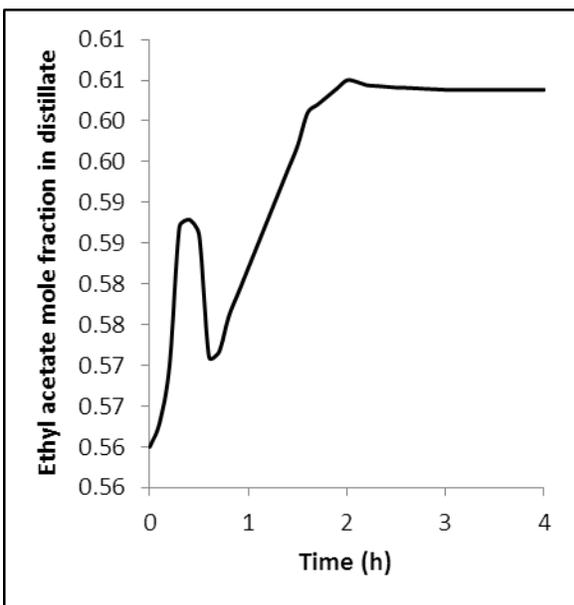
$$V_p = \text{Max}(\text{Min}(V_{pt}, 1), 0)$$

(11)

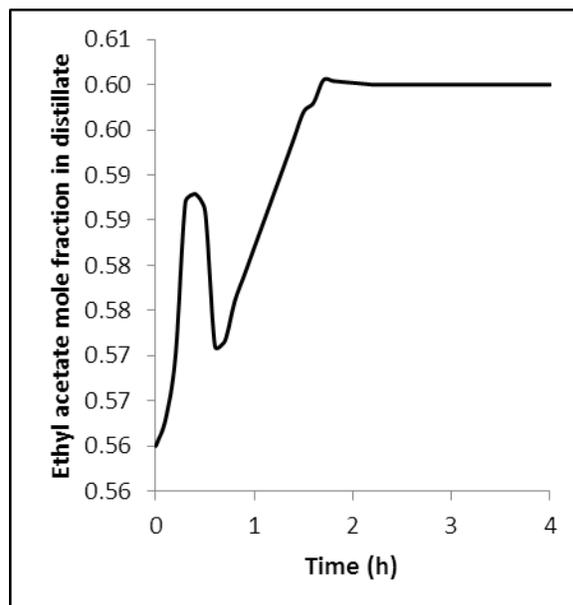
*ISE* is the integral square of the controller error and its magnitude indicates the performance of the controller. Therefore by minimizing the integral square error the optimal performance of the controller is obtained. The tuning of controller parameters  $K_c$  and  $\tau_i$  to minimise *ISE* was implemented by carrying out an optimization exercise with the objective being to minimise the integral square error. The initial guesses of the controller parameters were obtained by the Ziegler-Nichols (ZN) tuning criterion [25]. The optimal value of *ISE* obtained was 587.85 and the corresponding values of controller parameters  $K_c$  and  $\tau_i$  were 0.13 and 7.05 respectively.

## **ETHYL ACETATE COMPOSITION CONTROL**

Because disturbances are usually introduced along with inlet streams, it is necessary to adopt a suitable control scheme in order to maintain feasible operation of the process under consideration. For the current study, the composition of ethyl acetate in the distillate was controlled by manipulating condenser heat duty such that liquid is vented to attain the set point of minimum ethyl acetate mole fraction of 0.60. In tuning the controller parameters, no set of controller parameters ( $K_c$  and  $\tau_i$ ) could be found that resulted in feasible operation of the process. This was because the solution of the optimisation problem formulated for tuning the controller parameters resulted in huge constraint violations. Figures 10 and 11 show the response of ethyl acetate composition in the distillate to control action at different values of proportional gain.



**Figure 10:** Response of ethyl acetate composition in the distillate to control action when  $kc=0.5$



**Figure 11:** Response of ethyl acetate composition in the distillate to control action when  $kc=1.0$

Both Figs. 10 and 11 show that the composition of ethyl acetate reaches a steady value after some time. Figure 10 shows that using a value of 0.5 for proportional gain, the composition of ethyl acetate in the distillate reaches a steady set point value of 0.6 after about 2 hours of simulated time. Using a value of 1.0 for proportional gain, the composition of ethyl acetate in the distillate as shown in Fig. 11 reaches a steady set point value of 0.6 after about 3 hours of simulated time. The results obtained here agrees with the findings of Kumar<sup>[24]</sup> where they reported stable controlled ethyl acetate composition of not more than 0.60.

## CONCLUSIONS

Modelling, simulation and control of an ethyl acetate producing reactive distillation column system was accomplished using commercially available process simulation software, CHEMCAD. Results obtained by simulating the process at steady state show that double feeding strategy gives ethyl acetate with a higher purity when compared to single feeding strategy. This agrees with the findings of Vora & Daoutidis<sup>[14]</sup> who also worked on the same system and reported that top product purity can be improved by feeding reactants in different trays. The optimum location of feed stage was such that acetic acid is fed to stage 2 while ethanol is fed to stage 12. A reflux ratio of 12 resulted in optimum composition of ethyl acetate in the liquid phase and increasing the distillate rate resulted in an increase in the composition of ethyl acetate in the liquid phase.

Based on dynamic simulations of the reactive distillation column system under consideration, reboiler pressure control was achieved by manipulating the flow rate of steam to the reboiler. Control of the purity of ethyl acetate in the distillate was achieved by manipulating condenser heat duty such that liquid is vented to attain the set point of minimum ethyl acetate mole fraction of 0.60. Tuning of controller parameters was achieved using the Ziegler-Nichols (ZN) tuning criterion [25]. An optimal value of integral square error (*ISE*) of 587.85 and corresponding values of 0.13 and 7.05 for proportional gain and integral time constant ( $K_c$  and  $\tau_I$ ) respectively

## **NOMENCLATURE**

$A_p$	Pre exponential factor for the <i>p</i> th term
$C$	Number of components
$E_i$	Residual energy for stage <i>i</i>
$E_p$	Activation energy of the reaction for the <i>p</i> th term
$f_{i,j}$	Liquid component molar flow for component <i>j</i> in fresh feed to stage <i>i</i>
$g_{i,j}$	Vapour component molar flow for component <i>j</i> in fresh feed to stage <i>i</i>
$h_i$	Enthalpy of the primary liquid leaving stage <i>i</i>
$H_i$	Enthalpy of the primary vapour leaving stage <i>i</i>
$H_{Fi}$	Liquid enthalpy of feed stream on stage <i>i</i>
$H_{Gi}$	Enthalpy of total vapour phase on stage <i>i</i>
$K_{i,j}$	VLE ratio for component <i>j</i> on stage <i>i</i>
$k_p$	Reaction rate constant for the <i>p</i> th term
$l_{i,j}$	Moles of component <i>j</i> in the primary liquid leaving stage <i>i</i>
$f_{i,j}$	Residual component material balance for component <i>j</i> on stage <i>i</i>
$N$	Number of stages
$NR$	Number of reactions
$Q_i$	Heat duty at stage <i>i</i>
$R$	Reflux ratio
$S_i$	Ratio of vapour draws off to primary vapour on stage <i>i</i>
$T_i$	Temperature on stage <i>i</i>
$U_i$	Liquid hold up on stage <i>i</i>
$\nu_{j,n}$	Stoichiometric coefficient of component <i>j</i> in reaction <i>n</i>
$v_{i,j}$	Moles of component <i>j</i> in the primary vapor leaving stage <i>i</i>
$x_j$	Liquid phase mole fraction of component <i>j</i>
$y_j$	Vapour phase mole fraction of component <i>j</i>

- $x_{i,j}$  Liquid phase mole fraction of component  $j$  on stage  $i$   
 $y_{i,j}$  Vapour phase mole fraction of component  $j$  on stage  $i$

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